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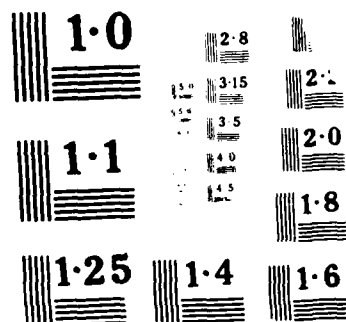
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## A Comparison of Various Alkali Gas Cell Atomic Frequency Standards

J. C. CAMPARO and R. P. FRUEHOLZ  
Chemistry and Physics Laboratory  
The Aerospace Corporation  
El Segundo, CA 90245

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Michael J. Mitchell

MICHAEL J. MITCHELL, Lt, USAF  
MOIE Project Officer  
SD/CWNZS

Raymond M. Leong

RAYMOND M. LEONG, Major, USAF  
Deputy Director, AFSTC West Coast Office  
AFSTC/WCO OL-AB

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19. ABSTRACT (Continue on reverse if necessary and identify by block number)  In this study a theoretical comparison among various alkali gas cell atomic frequency standards is undertaken, specifically: $Rb^{85}$ , $Rb^{87}$ , $Cs^{133}$ , $Cs^{135}$ and $Cs^{137}$ . It is found that $Rb^{87}$ exhibits the best potential shot-noise-limited performance of all the candidate alkalis (in a minimum volume TE <sub>111</sub> microwave cavity $Q(\tau) = 5.4 \times 10^{-15}/\sqrt{\tau}$ ), and that this is due to: 1) a large ground-state hyperfine splitting, and 2) a low value of nuclear spin. Additionally, the calculations indicate the importance of microwave cavity geometry on the stability one can attain with a gas cell atomic frequency standard.					
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## 1. INTRODUCTION

Though gas cell atomic frequency standards based on various alkalis have been discussed in the past (Refs. 1-5), notably the  $\text{Cs}^{133}$  gas cell standard [3][4], none has ever come near to rivalling the popularity of the  $\text{Rb}^{87}$  gas cell standard. Obviously, there are several factors which contribute to this situation; for example,  $\text{Rb}^{87}$  has the second largest ground-state hyperfine splitting of all the stable alkali isotopes (implying a relatively high atomic Q), and it has a low value of nuclear spin I so that optical pumping is relatively efficient in populating the  $m_F = 0$  state. The primary reason for the supremacy of the  $\text{Rb}^{87}$  standard, though, lies with the fortuitous overlap of  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  optical absorption lines, which allows for the construction of a very simple and efficient hyperfine filter for optical pumping with alkali discharge lamps (Ref. 6). However, with the advent of narrowband lasers for optical pumping in atomic frequency standards (Refs. 7, 8), especially single-mode diode lasers (Ref. 9), the prerequisite of an efficient hyperfine filter in the design of a gas cell standard has been eliminated. This in turn has diminished the intrinsic attractiveness of a  $\text{Rb}^{87}$  gas cell standard. In particular, given the 35% greater ground state hyperfine splitting of  $\text{Cs}^{133}$ , one must seriously question the relative merits of a rubidium standard in the absence of a hyperfine filter requirement. In the present study it is therefore our desire to theoretically explore the shot-noise limited performance of various laser pumped alkali gas cell standards, so that their intrinsic performance capabilities can, to a degree, be assessed.

Considering the fact that there are very many alkali isotopes with various values of nuclear spin and various ground state hyperfine splittings, it is necessary to reduce this host of candidate gas cell standards by requiring the candidate alkali isotope to possess a few reasonable characteristics. Obviously, the first requirement is that the alkali isotope be either stable or long lived. Specifically, we must require that the isotope have a half-life greater than ten years. This is particularly

important for space applications of frequency standards, where longevity is of prime importance. Furthermore, if major redesign of the standard is to be avoided, the alkali isotope must have half-integer nuclear spin, so that the standard can be based on a field insensitive 0-0 transition (Ref. 10). Thus, considering a full range of alkali isotopes (Ref. 11), one is left with the series of nine candidate alkalis listed in Table I. Additionally, in order to obtain efficient laser pumping, one would require that the hyperfine resonances be well resolved optically, that is, that the Doppler broadening be less than the ground-state hyperfine splitting. Consequently, out of the host of original candidate alkalis, only the following five will be considered for further comparison (Ref. 12):  $\text{Rb}^{85}$ ,  $\text{Rb}^{87}$ ,  $\text{Cs}^{133}$ ,  $\text{Cs}^{135}$  and  $\text{Cs}^{137}$ .

Table I. Some Properties of the Candidate Alkali Isotopes.

ALKALI	NUCLEAR SPIN I	% ABUNDANCE OR HALF-LIFE	DOPPLER BROADENING (MHz)*	HYPERFINE SPLITTING (MHz)
$7_{\text{Li}}$	$3/2$	92.58%	2340	804
$23_{\text{Na}}$	$3/2$	100%	1470	1772
$39_{\text{K}}$	$3/2$	93.10%	870	462
$41_{\text{K}}$	$3/2$	6.88%	850	254
$85_{\text{Rb}}$	$5/2$	72.15%	580	3036
$87_{\text{Rb}}$	$3/2$	27.85%	570	6835
$133_{\text{Cs}}$	$7/2$	100%	420	9193
$135_{\text{Cs}}$	$7/2$	$3 \times 10^6$ yr	420	9724
$137_{\text{Cs}}$	$7/2$	30.0 yr	420	10,116

\* Calculated Assuming a Vapor Temperature of 100°C



## II. OVERVIEW OF THE GAS CELL FREQUENCY STANDARD MODEL AND CALCULATION

In previous publications (Refs. 13, 14) we have discussed a non-empirical model of the gas cell atomic frequency standard. Based on a specific gas cell geometry, the model calculates the standard's anticipated frequency stability in terms of the positive value of the square root of the Allan variance, the quantity we refer to as the "Allan deviation." Calculation of the amount of optical pumping radiation transmitted through the atomic vapor storage cell as a function of injected microwave signal frequency, yields an atomic hyperfine lineshape. The derivative of this lineshape is proportional to the "error signal" upon which the operation of the standard is based (Refs. 1-5). For these calculations, the only noise source considered is the shot noise generated at the photodiode used to detect the transmitted light. This noise is considered the principal limitation to gas cell standard short-term frequency stability. Its exclusive consideration not only leads to accurate performance modelling, but also results in a consistent estimate of the performance of all the potential standards that are analyzed. To perform the analysis, the model considers the relevant gas phase physics as occurring on two different scales. On what we term the "microscopic scale" the 0-0 hyperfine transition lineshape of an arbitrary alkali atom of half-integer nuclear spin is determined by the generalized Vanier theory of alkali atom hyperfine optical pumping (Refs. 15, 16). Among other parameters, this theory considers the dependence of the hyperfine lineshape on optical pumping light intensity and microwave Rabi frequency. However, because the buffer gas pressure in a gas cell standard effectively freezes the alkali atoms in place on time scales of the order of a Rabi period (Ref. 17), and because the alkali vapor is not necessarily optically thin; these two parameters, and hence the microscopic lineshape, vary from atom to atom within the vapor. Furthermore, as a result of diffusion to the resonance cell walls, where the atoms immediately depolarize on impact, there is a spatial distribution of hyperfine polarization  $\langle \vec{I} \cdot \vec{S} \rangle$ . In one sense, this spatial distribution of  $\langle \vec{I} \cdot \vec{S} \rangle$  can be imagined as being superimposed on the microscopic physics. Thus, there is also a "macroscopic scale" of physics in the problem which is related to the

spatial variation of: (a) the optical pumping light intensity, (b) the microwave Rabi frequency, and (c)  $\langle \vec{I} \cdot \vec{S} \rangle$  as a result of diffusion to the resonance cell walls.

In order to treat this macroscopic scale of physics in a reasonably lucid manner, the problem is reduced to one dimension, so that only the longitudinal variation of the optical pumping rate and microwave field strength is considered. This is reasonable because the microwave field can be made uniform in the transverse dimension by dielectrically loading the cavity (Ref. 18), and because the laser intensity can easily be made uniform across the face of the resonance cell. The microwave Rabi frequency distribution along the axial dimension is determined by the microwave cavity mode, assuming that the atomic resonance cell fills the microwave cavity. The axial variation of the optical pumping light intensity is determined by computing a "global" optical pumping parameter  $\zeta$  in a self-consistent manner. In essence, this global optical pumping parameter determines the fractional population in the optically absorbing hyperfine multiplet, and thus the optical depth of the vapor as a result of optical pumping. Since the model assumes that the alkali atoms are effectively frozen in place in the resonance cell, the first order change in transmitted light intensity as a function of microwave Rabi frequency, for a uniform slice of vapor of thickness  $dz$ , depends on the local values of the optical pumping light intensity and microwave Rabi frequency. In order to include the effect of axial diffusion, this first order macroscopic solution is multiplied by the envelope function  $f(z)$ , which describes the axial distribution of hyperfine polarization in an optically thin vapor. (This procedure for treating diffusion is discussed more fully in Ref. 13.) When considering optical pumping with lamps, where the relative optical pumping rates are typically low, it is fair to approximate  $f(z)$  by Minguizzi et al.'s first order diffusion mode (Ref. 19)

$$f(z) = \sin(\pi z/L), \quad (1)$$

where  $L$  is the length of the resonance cell. However, as previously discussed (Ref. 14), when the optical pumping light source is a laser, the envelope

function must be generalized so as to be valid for arbitrary optical pumping rates

$$f(z) = \frac{[1 - \exp(-\alpha z)] [\exp(\alpha L) - \exp(\alpha z)]}{[1 - \exp(-\alpha L/2)] [\exp(\alpha L) - \exp(\alpha L/2)]} \quad (2)$$

In the present analysis, we consider an alkali gas cell atomic frequency standard operating in its traditional configuration (i.e., cw optical pumping), except that the typical rf discharge lamp used for optical pumping is replaced by a single-mode laser tuned to the  $D_2$  optical absorption resonance of the alkali [ $n^2P_{3/2} - n^2S_{1/2}(F=I+1/2)$ ]. Furthermore, we assume that there is no filter cell, and that the resonance cell contains a pure isotopic vapor of the alkali under consideration. The calculations are performed in such a way that, for a particular incident laser intensity, we calculate the resonance cell temperature and peak microwave Rabi frequency that minimize the shot-noise-limited Allan variance as well as the minimum Allan variance value itself. Temperature enters the calculations through: 1) the temperature sensitivity of the alkali's diffusion coefficient, 2) the collision frequency of the alkali atoms (i.e., the relative speed of the atoms), and 3) the alkali vapor number density which determines both the spin exchange rate and the vapor's optical depth. Additionally, we consider a clock operating with a  $TE_{111}$  cylindrical microwave cavity of minimum volume

$$L = c \sqrt{3} / 2\nu_0 \quad (3a)$$

$$R = 1.841 \left[ \left( \frac{2\pi\nu_0}{c} \right)^2 - \left( \frac{\pi}{L} \right)^2 \right]^{-1/2} \quad (3b)$$

where  $R$  is the cavity's radius, and  $\nu_0$  is the alkali's 0-0 hyperfine transition frequency. Thus, alkali gas cell standards with high hyperfine transition frequencies are modelled as operating with appropriately smaller microwave cavities and resonance cells. Other general parameters used in the calculations are presented in Table II.

Table II. Parameters Used in the Calculation of the Clock Signal Shot Noise.

PARAMETER	VALUE
Laser Linewidth	50 MHz
Nitrogen Buffer Gas Pressure	10 torr
Photodetector Responsivity	0.5 amps/watt

### III. RESULTS

The basic results of the calculations are presented in Fig. 1 and Table III. Clearly,  $\text{Rb}^{87}$  exhibits the best shot-noise-limited performance ( $\sigma_y(\tau) \approx 5.4 \times 10^{-15}/\sqrt{\tau}$ ), and is roughly a factor of three better than any of the cesium isotopes. However, before saying too much about this result, one should note that the shot-noise-limited performance of the three cesium isotopes is a decreasing function of the isotope's hyperfine transition frequency. Given the equality of the nuclear spins of these isotopes, and the fact that the atomic line  $Q$  increases proportionate to  $\nu_0$ , the result is counter-intuitive; typically, we would expect  $\sigma_y(\tau) \sim 1/\nu_0$  under the condition of equal isotope nuclear spin.

Table III. Best Allan Deviations (i.e., Square Root of the Allan Variance) for the Various Candidate Alkalies as Well as the Corresponding Laser Intensity, Cell Temperature, and Peak Microwave Rabi Frequencies.

ALKALI	$\sigma_y^{\text{best}}(\tau)$	LASER INTENSITY $\mu\text{W}/\text{cm}^2$	TEMPERATURE $^{\circ}\text{C}$	PEAK RABI FREQUENCY Hz	MINIMUM CAVITY VOLUME $\text{cm}^3$
$85\text{Rb}$	$5.5 \times 10^{-15}/\sqrt{\tau}$	90	67	97	338.0
$87\text{Rb}$	$5.4 \times 10^{-15}/\sqrt{\tau}$	128	73	155	29.6
$133\text{Cs}$	$1.3 \times 10^{-14}/\sqrt{\tau}$	371	79	380	12.2
$135\text{Cs}$	$1.4 \times 10^{-14}/\sqrt{\tau}$	291	78	309	10.3
$137\text{Cs}$	$1.4 \times 10^{-14}/\sqrt{\tau}$	315	79	346	9.1

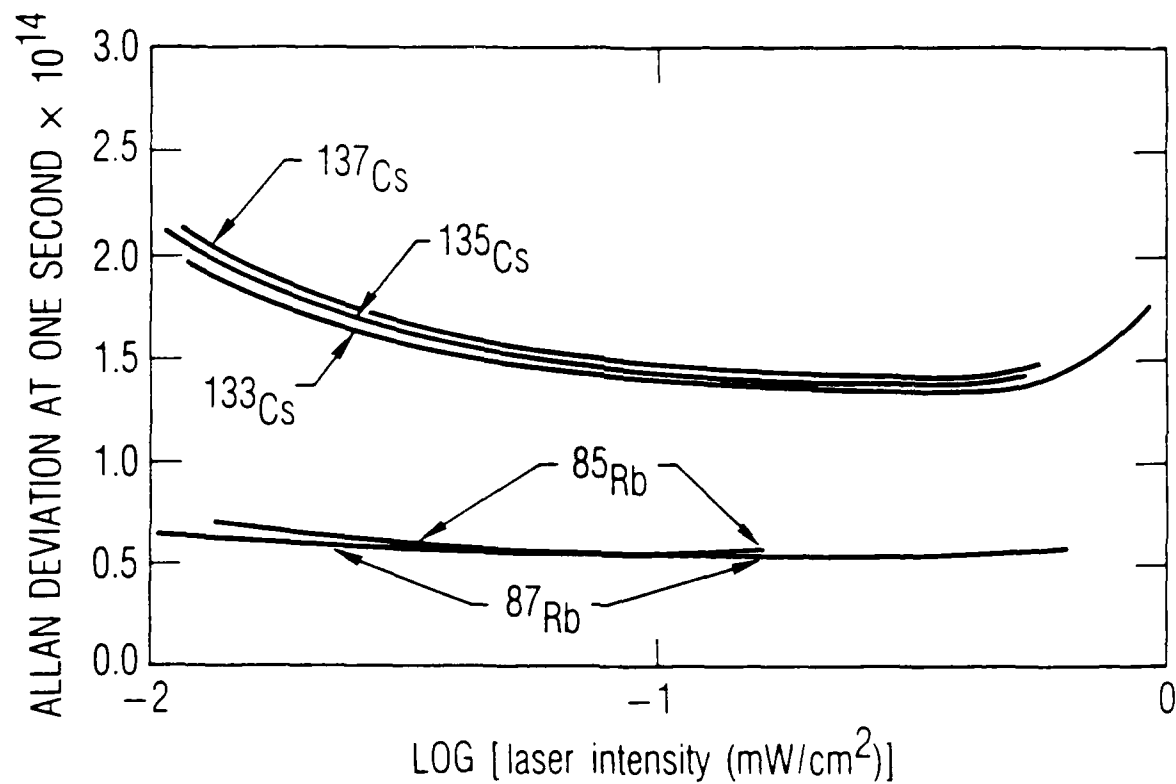


Fig. 1. Various Alkali Isotope Curves Showing the best Allan Deviations (i.e., Square Root of Allap Variance) as a Function of Incident Laser Intensity. Note that a  $\text{Rb}^{87}$  standard shows the best predicted performance. As discussed in the text the relation between the cesium isotope performances reflects the influence of cavity geometry on the Allan deviation, as well as intrinsic difference between the isotopes.

The resolution of this apparent paradox resides in the fact that these calculations assume a minimum volume microwave cavity, and hence a minimum volume resonance cell. As can be seen with the aid of Eq. (3) the cell volume decreases like  $1/v_0^3$ . Thus, the cesium isotope results can be explained by postulating a negative correlation between microwave cavity size and the shot-noise-limited Allan variance. This hypothesis has been substantiated by running the cesium isotope calculations for the fictitious case of equal cavity radii and lengths. The resulting Allan deviations scaled like  $1/v_0$  (i.e., Cs<sup>137</sup> showed a 10% improvement in shot-noise-limited performance compared to Cs<sup>133</sup>). Consequently, the results presented in Fig. 1 and Table III not only represent the intrinsic performance capabilities of the various alkali isotopes, but also geometrical effects associated with the microwave cavity's size.

How the cavity geometry affects gas cell frequency standard performance is not understood at the present time. One possible explanation is that by increasing the microwave cavity length, and hence the resonance cell length, one obtains good transmitted light signal amplitudes at relatively low alkali densities. Since low alkali densities imply low spin-exchange rates, and hence relatively higher atomic Qs and signal-to-noise ratios, longer resonance cells might be expected to exhibit improved clock performance. In the same vein, because larger resonance cell radii imply lower phenomenological diffusional relaxation rates, one again might expect some improvement in predicted clock stability with larger radii resonance cells. An alternative explanation, however, is derived from the axial distribution of hyperfine polarization and microwave field strength in the resonance cell. Perhaps the influence of these distributions on clock signal amplitude is such that longer resonance cells yield relatively higher signal levels, and hence improved clock stability. In order to differentiate among these and other potential explanations for the influence of microwave cavity geometry on clock stability, additional calculations need to be performed, preferably with a more rigorous 3-dimensional model of the gas cell atomic frequency standard.

To mitigate the influence of cavity geometry on the calculation of shot-noise-limited performance, we considered the fictitious case of a Cs<sup>133</sup> gas

cell frequency standard operating with a  $TE_{111}$  microwave cavity having the same radius and length as a  $Rb^{87}$   $TE_{111}$  resonant cavity. Though the result of this calculation showed an improvement in clock stability compared to the  $Cs^{133}$  result given in table III, the best Allan deviation was nonetheless a factor of 1.6 larger than the corresponding Allan deviation of  $Rb^{87}$ . It would thus appear that the approximate factor of two difference in nuclear spins between  $Rb^{87}$  and  $Cs^{133}$  (i.e., the factor of two difference in the ground-state degeneracies) more than compensates for the 35% difference in the alkali ground state hyperfine splittings.

It is also surprising that  $Rb^{85}$  should display such good performance compared to  $Rb^{87}$ , considering that it has half the hyperfine resonance frequency and a 50% greater ground state degeneracy resulting from its larger nuclear spin. In light of the preceding discussion of the Cs isotope Allan variances, we attribute this relatively good performance to  $Rb^{85}$ 's ten times larger minimum microwave cavity volume. This again emphasizes the influence of the physics package geometry on the ultimate shot-noise-limited performance that can be attained with the gas cell standard.



#### IV. SUMMARY

The present calculations indicate that a  $\text{Rb}^{87}$  gas cell standard shows the greatest potential for frequency stability, and in this regard nature has been uncommonly propitious. One should not, however, interpret this result as a superiority of the  $\text{Rb}^{87}$  standard in all regards. For example, if it is of primary importance to construct a miniature gas cell standard, then  $\text{Cs}^{133}$  might prove to be more advantageous given the fact that its minimum volume cavity occupies less than half the volume of a corresponding  $\text{Rb}^{87}$  cavity. Additionally, magnetic field sensitivities are less for  $\text{Cs}^{133}$  as a consequence of its greater hyperfine transition frequency. The only statement one should make regarding the present results is that of all the possible alkali gas cell standards that could be considered, a  $\text{Rb}^{87}$  standard appears to yield the best attainable shot-noise-limited performance.

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$$\nu_{mn} = \nu_0 + \frac{KB}{[I]} (m+n) + \frac{(KB)^2}{2 \nu_0} \left( 1 - \frac{(m^2 + n^2)}{2 [I]^2} \right)$$

where  $\nu_0$  is the unperturbed hyperfine splitting,  $K = g \mu_B / h = 2.80$  MHz/gauss,  $[I] = (2I+1)$ , and  $B$  is the magnitude of the static magnetic field. Clearly the transitions which, to first order, are insensitive to the magnetic field correspond to transitions where  $m = -n$ . However, as a consequence of the magnetic dipole selection rules, not all combinations of  $m$  and  $n$  yield allowed transitions. Specifically, we have the requirement that  $\Delta m_F = m - n = 0, \pm 1$ . Consequently, for any alkali-like atom the only magnetic field insensitive transitions which are allowed correspond to: i)  $|a, 0\rangle - |b, 0\rangle$  or ii)  $|a, \pm 1/2\rangle - |b, \mp 1/2\rangle$ . Case i) corresponds to the well known 0-0 hyperfine transition occurring in half-integer species, while case ii) corresponds to alkali-like atoms with integer values of the nuclear spin. Therefore, one could base a gas cell clock on an integer nuclear spin alkali, but the stimulating microwave field would have to drive  $\Delta m_F = \pm 1$  transitions rather than the traditional clock's  $\Delta m_F = 0$  transition. To accomplish this would require a redesign of the microwave cavity configuration in the clock, and would present a series of difficulties and variations beyond the scope of the present study.

It should be noted, however, that from the above equation for  $\nu_{mn}$  one can show that the  $|a, \pm 1/2\rangle - |b, \mp 1/2\rangle$  transition frequencies are degenerate to second order in the magnetic field. Consequently, for a clock based on an  $I=1$  alkali-like atom, four out of the six ground state Zeeman sublevels would contribute to the clock signal. This could result in a relative improvement in the clock's signal to noise ratio, and hence the clock's short term stability. Suffice it to say that gas cell frequency standards based on integer nuclear spin species may have potentially significant attributes.

We wish to thank the referee for pointing out the fact that integer nuclear spin alkalis can have field insensitive transitions, which stimulated us into rethinking the question of gas cell standards based on integer nuclear spin species.

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## LABORATORY OPERATIONS

The Aerospace Corporation functions as an "architect-engineer" for national security projects, specializing in advanced military space systems. Providing research support, the corporation's Laboratory Operations conducts experimental and theoretical investigations that focus on the application of scientific and technical advances to such systems. Vital to the success of these investigations is the technical staff's wide-ranging expertise and its ability to stay current with new developments. This expertise is enhanced by a research program aimed at dealing with the many problems associated with rapidly evolving space systems. Contributing their capabilities to the research effort are these individual laboratories:

Aerophysics Laboratory: Launch vehicle and reentry fluid mechanics, heat transfer and flight dynamics; chemical and electric propulsion, propellant chemistry, chemical dynamics, environmental chemistry, trace detection; spacecraft structural mechanics, contamination, thermal and structural control; high temperature thermomechanics, gas kinetics and radiation; cw and pulsed chemical and excimer laser development including chemical kinetics, spectroscopy, optical resonators, beam control, atmospheric propagation, laser effects and countermeasures.

Chemistry and Physics Laboratory: Atmospheric chemical reactions, atmospheric optics, light scattering, state-specific chemical reactions and radiative signatures of missile plumes, sensor out-of-field-of-view rejection, applied laser spectroscopy, laser chemistry, laser optoelectronics, solar cell physics, battery electrochemistry, space vacuum and radiation effects on materials, lubrication and surface phenomena, thermionic emission, photo-sensitive materials and detectors, atomic frequency standards, and environmental chemistry.

Computer Science Laboratory: Program verification, program translation, performance-sensitive system design, distributed architectures for spaceborne computers, fault-tolerant computer systems, artificial intelligence, microelectronics applications, communication protocols, and computer security.

Electronics Research Laboratory: Microelectronics, solid-state device physics, compound semiconductors, radiation hardening; electro-optics, quantum electronics, solid-state lasers, optical propagation and communications; microwave semiconductor devices, microwave/millimeter wave measurements, diagnostics and radiometry, microwave/millimeter wave thermionic devices; atomic time and frequency standards; antennas, rf systems, electromagnetic propagation phenomena, space communication systems.

Materials Sciences Laboratory: Development of new materials: metals, alloys, ceramics, polymers and their composites, and new forms of carbon; non-destructive evaluation, component failure analysis and reliability; fracture mechanics and stress corrosion; analysis and evaluation of materials at cryogenic and elevated temperatures as well as in space and enemy-induced environments.

Space Sciences Laboratory: Magnetospheric, auroral and cosmic ray physics, wave-particle interactions, magnetospheric plasma waves; atmospheric and ionospheric physics, density and composition of the upper atmosphere, remote sensing using atmospheric radiation; solar physics, infrared astronomy, infrared signature analysis; effects of solar activity, magnetic storms and nuclear explosions on the earth's atmosphere, ionosphere and magnetosphere, effects of electromagnetic and particulate radiations on space systems; space instrumentation.

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